Coexistence of glassy antiferromagnetism and giant magnetoresistance (GMR) in Fe/Cr multilayer structures

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Using temperature-dependent magnetoresistance and magnetization measurements on Fe/Cr multilayers that exhibit pronounced giant magnetoresistance (GMR), we have found evidence for the presence of a glassy antiferromagnetic (GAF) phase. This phase reflects the influence of interlayer exchange coupling (IEC) at low temperature ($T < 140$K) and is characterized by a field-independent glassy transition temperature, T_g, together with irreversible behavior having logarithmic time dependence below a "de Almeida and Thouless" (AT) critical field line. At room temperature, where the GMR effect is still robust, IEC plays only a minor role, and it is the random potential variations acting on the magnetic domains that are responsible for the antiparallel interlayer domain alignment.

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Given the established presence of GMR-based devices in technology, especially in the multi-billion dollar computer hard disk drive market, it may come as a surprise that there is still an incomplete scientific understanding of the GMR effect[1]. The mechanism for GMR, first observed in single crystalline (100) Fe/Cr multilayers grown by molecular beam epitaxy[2, 3] and subsequently in magnetron-sputtered polycrystalline films[4], relies on spin-dependent scattering[5] and the associated dependence of resistance on the relative orientations of the magnetizations in neighboring layers. It is important to recognize that interlayer exchange coupling (IEC) is not necessarily required for a GMR effect[1]. In a particularly simple manifestation, two neighboring films, separated by a non-magnetic spacer layer, could have different coercive fields, thus giving rise to antiparallel alignment and a GMR effect, as the external field is cycled[6]. Randomness[7, 8] and competing interactions such as biquadratic coupling[9, 10] can also play a significant role. In this paper we identify a glassy antiferromagnetic (GAF) phase which by marking the influence of IEC at low temperatures implies that at higher temperatures random potential variations rather than IEC are responsible for antiparallel alignment.

Our Fe/Cr multilayer samples have been prepared on silicon substrates by ion beam sputter deposition of separate Fe and Cr targets. Extensive characterization of the deposited multilayers showed distinct compositional and structural modulations with well-defined interfaces and a surface roughness on the order of 5Å. Ten and thirty-layer stacks with the repeat sequence [Fe(20Å)/Cr(d_{Cr})] are typically deposited and passivated with a 50Å-thick Cr layer. The Cr spacer thickness d_{Cr} is varied over the range 8–12Å. The inset of Fig. 1 shows typical GMR traces at 300K and 10K for the magnetic field parallel to the planes of a [Fe(20Å)/Cr(12Å)]×30 sample.

In Fig. 1 we show a selected subset of temperature-dependent field-cooled (FC, open symbols) and zero-field-cooled (ZFC, closed symbols) magnetization data for a thirty layer sample with $d_{Cr} = 12\AA$ and a GMR ratio ($(R(0) - R(H))/R(0)$, Fig. 1 inset) of 20.6% at 10K. The data were taken using a SQUID magnetometer in fields (indicated on the plot) oriented parallel to the layers. At each field the corresponding FC and ZFC curves

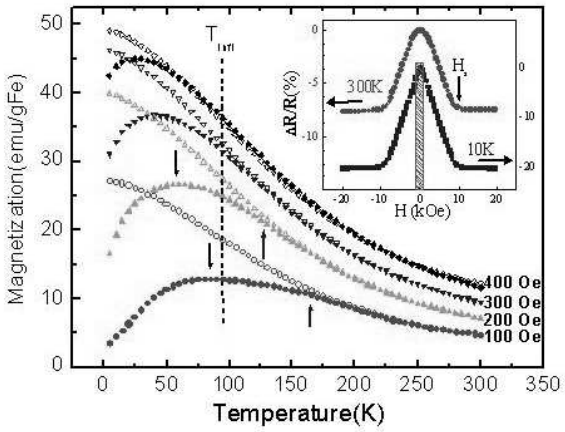

FIG. 1: Magnetization of a multilayer sample ([Fe(20Å)/Cr(12Å)]×30) normalized to the weight of iron plotted as a function of temperature at the indicated fields. The data at each field are taken in pairs: the open(solid) symbols referring to the field-cooled, FC, (zero-field cooled, ZFC) procedure. The vertical arrows and dashed line are described in the text. Inset, dependence of the giant magnetoresistance (GMR) ratio on applied field for the same film at 300K (left axis) and at 10K (right axis).

can be characterized by three distinct temperatures: an irreversibility temperature $T_{irr}(H)$ denoting the bifurcation point below which there is hysteresis (upward arrows), a temperature $T_m(H)$ (downward arrows) denoting the maximum in each of the ZFC curves, and an inflection temperature T_{infl} (vertical dashed line) which marks the inflection point of each FC curve. Evidently T_{infl} is quite robust and independent of field, having a value $T_{infl} = 93.0 \pm 1.4K$ determined to relatively high precision from FC measurements at 5 different fields spanning the range 50-400 Oe.

Compelling evidence for an interlayer rather than intralayer effect is found in the resistance measurements of Fig. 2 on the same sample. For each datum on this graph, the sample was zero-field cooled to the target temperature, the resistance $R(0)$ measured, and then a field applied to measure the change in resistance $\delta R = R(0) - R(H)$. The ratio $\mid \delta R/R(0) \mid$ is plotted against temperature for the fields indicated in the legend. The striking aspect of these data is that although the peaks are not as pronounced as those in the ZFC magnetizations of Fig. 1, their positions in an H-T plot of Fig. 3 (open triangles) show close similarity with respect to the positions of the ZFC peaks (solid circles).

The presence of a spin-glass-like phase is buttressed by our finding that $T_m(H)$ defines a critical field line (solid circles in Fig. 3) which delineates the onset of strongly irreversible behavior and has the de Almeida and Thouless (AT) form[11, 12], $H/T \propto (T_g/T - 1)^{3/2}$ (inset), where T_g is the spin glass temperature. Although other criteria could have been used[12], we note that our choice of $T_m(H)$ as the criterion determining the AT line has particular cogency because it obeys the scaling form of the AT prediction and extrapolates at zero field to a field-independent glass temperature $T_g = 1.51 \times T_{infl} = 140$K, where T_{infl}, an apparent fixed point, has been independently determined from the FC data (dashed line of Fig. 1).

An additional and essential ingredient for a glassy phase is the presence of disorder measured by the variance, ΔJ, in the antiferromagnetic (AF) coupling strengths. This variance arises because of the existence of domains and the concomitant constraints imposed by intralayer dipolar interactions The exchange energy between two Fe moments separated by a spacer layer is of the form $E = J_{AF}\cos(\Psi)$, where Ψ denotes their relative angle. The intralayer domain structure imposes well-defined orientations of the spins and this constraint will not be consistent, in general, with $\Psi = \pi$ (i.e. with a minimum value of E). Because of the long-range nature of dipolar interactions, lowering the exchange energy requires the overturning of one or of several clusters of Fe moments, which is energetically inhibited at low temperature. In this regime, Ψ behaves like a pseudo random variable. A realistic estimate for ΔJ can be obtained by assuming a flat distribution for the values of Ψ on the [0, 2π] interval, leading to $\Delta J = J_{AF}/\sqrt{2}$. At $T > T_g$, IEC is present but ineffective because the intralayer dipolar interactions dominate.

Many glassy systems, including the one discussed here,

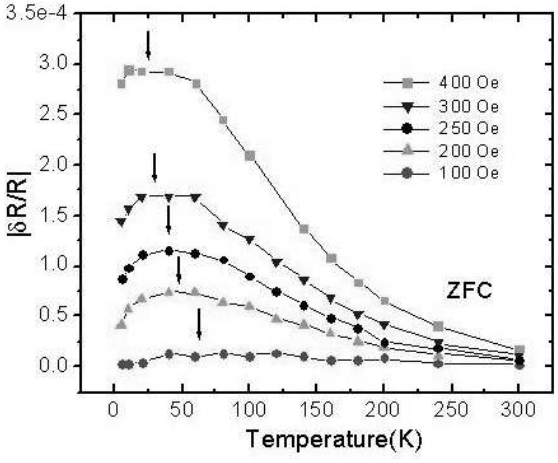

FIG. 2: Temperature dependence of the relative changes in resistance at the fields indicated in the legend for the same sample characterized in Fig. 1. For each data point, the sample was zero-field cooled as described in the text. The vertical arrows indicate the positions of the maxima for each field and define a critical field dependence similar to that defined by the maxima of the ZFC magnetizations in Fig. 1.

show AT like boundaries without being Ising spin glasses to which the theory[12, 13] strictly applies. The GAF phase associated with our GMR multilayers is clearly not an Ising system and is more reasonably described in terms of an anisotropic vector model in which the elemental spins, belonging to magnetized domains, are coupled ferromagnetically in the X-Y plane and antiferromagnetically in the perpendicular direction. For such vector glass systems there is an additional degree of freedom in the order parameter and the true phase boundary is delineated at higher temperatures and fields by the Gabay-Toulouse (GT) boundary[14]. A more comprehensive viewpoint that facilitates understanding of our experiment can be gleaned from the schematic phase diagram, shown in Fig. 4 for the H-T plane at $J_{AF}/\Delta J > 1$. (Note that the PM phase is not labeled as a ferromagnetic (FM) phase, since in the presence of a field there is no spontaneous symmetry breaking as the temperature is reduced through the Curie temperature.) In simplified terms the GT line (solid) can be thought of as denoting the onset of a phase transition to glassy behavior and the AT line (dotted) as the onset of pronounced irreversibility. (The experimental signature of the GT line, which has not been measured here, is a divergence in the transverse ac susceptibility.) At $H = 0$ both lines terminate at $T = T_g$.

The following three consequences, confirmed by experiment, are immediately apparent: Firstly, since $T_g \propto J_{AF}$ and $\Delta J \simeq J_{AF}$, it is clear that as T_g increases, the boundary of the GAF phase moves out to higher temperatures and fields. Experimentally this is confirmed in Fig. 3 where the AT line for the sample with $d_{Cr} = 10\mathring{A}$ (solid squares) has higher critical fields and a correspondingly higher T_g than the sample with $12\mathring{A}$ spacer. A second consequence is that the disorder-induced close proximity of T_g and J_{AF} implies that at low H the presence of an AF phase is obscured on the transition (Fig. 4, horizontal dashed arrow) from the PM to GAF phase. If this were not the case, then the field-cooled dc susceptibility would have a maximum at the AF boundary and then saturate at a smaller value as $T \to 0$. Such maxima are not observed! A third consequence supporting the existence of a GAF phase comes from the scaling of the field-cooled magnetization with H. Field- cooled (FC) magnetizations including those shown in Fig. 1 reveal that $M/H \sim H^{-u}$ as $T \to 0$. Here we find u=0.58(2) for 5K magnetization data taken at 7 different fields ranging from 100 to 800Oe, thus confirming behavior characteristic of spin glass systems below the lower critical dimension [12]. Finally, in addition to hysteresis, we also observe slow relaxations in the magnetization and resistance that are logarithmic in time and which, but for lack of space, can be explained by invoking constraints on the dynamics imposed by a hierarchy of domain sizes [15, 16].

To fully appreciate the role of randomness in multilayers, it is important to recognize the difference between

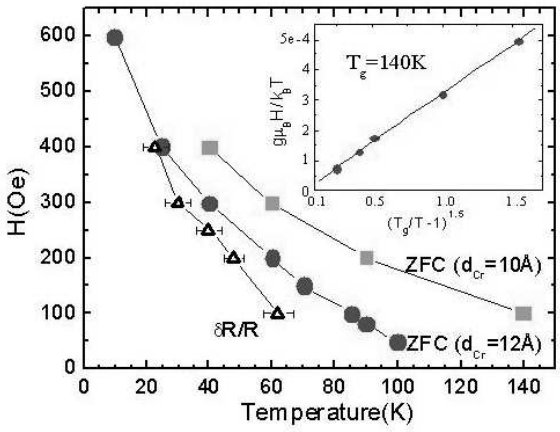

FIG. 3: Critical field lines for the 30 layer [Fe(20Å)/Cr(12Å)] (solid circles and open triangles) sample shown in Fig. 1 and for a second 30 layer [Fe(20Å)/Cr(10Å)] (solid squares) multilayer sample with smaller Cr spacer thickness. The solid symbols refer to determinations using the experimental $T_m(H)$'s of ZFC magnetizations and the open triangles are determined by similar peaks in the resistance measurements. Inset, plot of the high temperature points (solid circles) showing the de Almeida-Thouless (AT) scaling dependence for spin glasses.

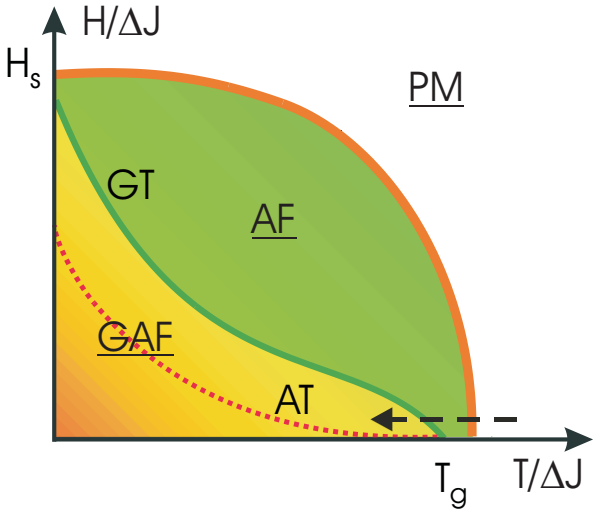

FIG. 4: Schematic of phase diagram in the H-T plane showing the relationship between the glassy antiferromagnetic (GAF), the antiferromagnetic (AF) and the paramagnetic (PM) phases. The axes are normalized as discussed in the text. The Gabay-Toulouse (GT) and de Almeida and Thouless (AT) line (dashed) are described in the text. For our samples the disorder is sufficiently large (i.e., $\Delta J \simeq J_{AF}$) and the field sufficiently low to ensure that the presence of an AF phase is obscured on the transition from the PM to GAF phase (horizontal dashed arrow).

GMR multilayers, in which there is a strong interaction between closely coupled interfaces, and bilayer or trilayer configurations in which such interactions can be ignored since there are at most only two interfaces. Thus for example, in studies of exchange bias in single ferromagnetic/antiferromagnetic (Co/CoO) bilayers[8], the onset of exchange bias, which is induced by random interactions[7], is observed to occur at a single temperature, the Neel temperature. By contrast, in our case there are two temperature ranges: $T < T_g = 140$K for glassiness and $T >\sim 250$K where there is a loss of AF order in Cr and disorder is still important. Accordingly, the picture described for FM/AF bilayers[8] is different for closely coupled multilayers where interactions between multiple ferromagnetic (FM) layers and interactions between interfaces should be taken into account. Similar considerations also apply to the magneto-optic Kerr effect (MOKE) and scanning electron microscopy with polarization analysis (SEMPA) studies[17] on Fe/Cr/Fe trilayers and magnetization and ferromagnetic resonance studies of CoFe/Mn/CoFe trilayers[10], all of which specialize to a specific type of spacer layer and do not include the multilayer interactions responsible for our GAF behavior. Our results are thus complementary yet distinct from the results of bilayer/trilayer experiments.

A consideration of the relevant energy scales and the mutual interactions of the magnetized domains in the Fe layers solidifies this emerging picture of spin-glass-like behavior in GMR multilayers. If adjacent Fe layers of thickness t and saturation magnetization M_s are coupled through an antiferromagnetic exchange J per unit area, then saturation at a field $H = H_s$ occurs when $J = HM_st/4$, a relation found by equating the field energy per unit area, HM_st, to the energy difference, $4J$, between the aligned and antialigned magnetic configurations . We note that a glass temperature near 140K corresponds to an antiferromagnetic coupling energy $\simeq 10$ meV, in good agreement with theoretical calculations[18, 19] for Fe/Cr layers. In the first calculation by Fishman and Shi[18] the Fe layers are exchange coupled below the Neel temperature T_n of the Cr spacer and a very strong AF coupling between the Fe and Cr moments at the interface is assumed. For our GAF phase T_n is in reality T_g. In the second calculation by Majumdar *et al.*[19] magnetoresistance data is well described by a theoretical expression in which RKKY interactions give a best fit AF coupling strength of (70 ± 20)K.

For $T > T_g$, the Fe layers are no longer AF coupled and the expression $J = HM_st/4$ to calculate the IEC is no longer relevant. In its place we use the expression[20, 21] $H_s = 4\pi M_s$, to calculate the maximum saturation field necessary to align dipolar-coupled domains within each layer. This expression is valid for both perpendicular and parallel fields[21]. The saturation fields of 10-20kOe in our samples (Fig. 1 inset) and similar samples reported by others[2, 4] are the right order of magnitude for Fe which with a saturation magnetization $M_s = 1700\text{Oe/cm}^3$ implies a maximum saturation field $H_s = 4\pi M_s = 21$kOe. For our three different samples with $d_{Cr} = 8$, 10 and 12$\mathring{A}$ we find a linear dependence of H_s on d_{Cr} which extrapolates to the origin ($d_{Cr} = 0$) to a value within 5% of H_s=21kOe, thus validating our use of this analysis.

To associate field scales with energy (or equivalently, temperature), we use the conversion ratio, $2.2\mu_B B/k_B T$=1.5T/K, where the magnetic moment of Fe is 2.2 Bohr magnetons. Accordingly, the dipolar interaction strengths measured by H_s, which are balanced by domain wall energies, are on the order of a few Kelvin and hence not strong enough at $T > T_g$ to determine domain orientation. Rather, domain orientation at $T > T_g$ is determined by the much stronger potential variations associated with crystalline anisotropies and the presence of impurities and defects. The presence of a GAF phase implies that IEC is effective in creating an anti-alignment effect beneficial to a large GMR effect only at low temperatures ($T < T_g$) and low fields ($H < H_{AT}$). The shaded region in the inset of Fig. 1 illustrates just how narrow this region is.

In summary, we show that a heretofore-unrecognized glassy antiferromagnetic (GAF) state coexists with GMR in polycrystalline Fe/Cr multilayer stacks. The very presence of this glassy phase sets an energy scale (T_g=140K) for antiferromagnetic interlayer exchange coupling (IEC) that is well below room temperature. We therefore conclude that, for temperatures greater than T_g, IEC plays only a minor role in forcing the antiparallel interlayer domain orientations that give rise to the ($H = 0$) high resistance state of multilayer Fe/Cr GMR samples. Rather, random potential variations, which constrain domain orientation, must be taken into account to understand GMR in multilayer GMR devices. The origin of the dependence of H_s on spacer thickness in multilayers as observed here and by others[2, 4] as well as the origin of the AF couplings for $T < T_g$ are totally open questions. This contrasts with the bilayer and trilayer cases[7, 8, 17] for which the AF couplings have a clear source.

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